

Work Function Lowering and Ponderomotive E-Acceleration for Ultra Low Emittance Beams

Károly Németh

Advanced Photon Source, Argonne National Laboratory

**In collaboration with Katherine C. Harkay (APS/ANL),
Michel van Veenendaal (APS/ANL & NIU), Linda Spentzouris
(APS/ANL & IIT), Jeff Terry (IIT), Marion White (APS/ANL),
Klaus Attenkofer (APS/ANL), Alexander Zholents (APS/ANL)
and George Srajer (APS/ANL)**

(based on K. Németh et.al, Physical Review Letters, 104, 046801 (2010))

Surface band of Cu(111)

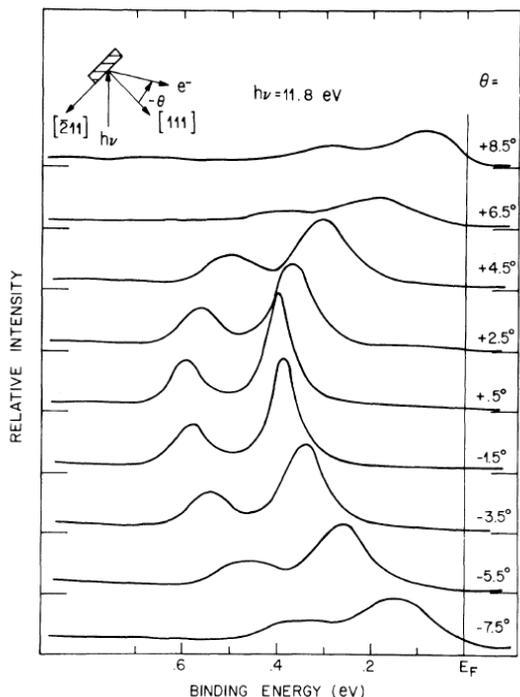


FIG. 1. Experimental energy distribution curves for the Cu(111) *sp* surface state for several angles near normal emission in the $\Gamma L X$ plane. The second peak is due to the Ar I doublet.

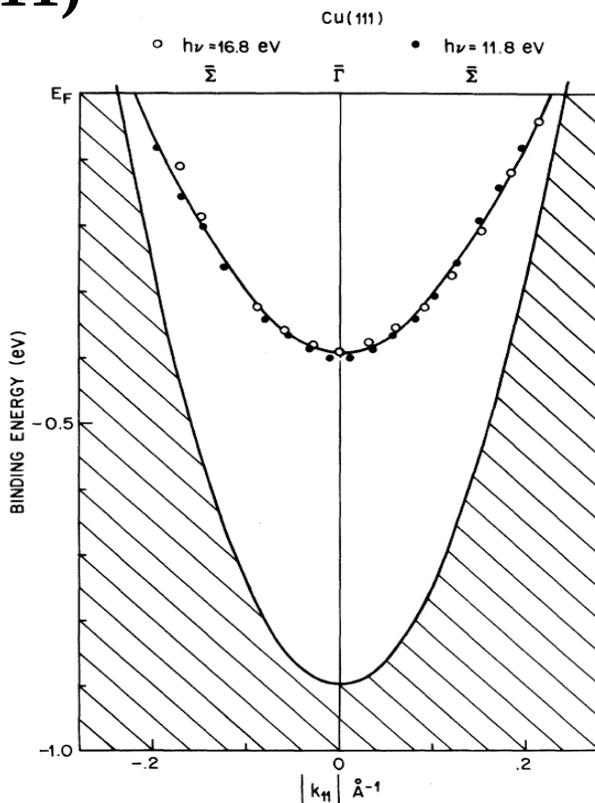
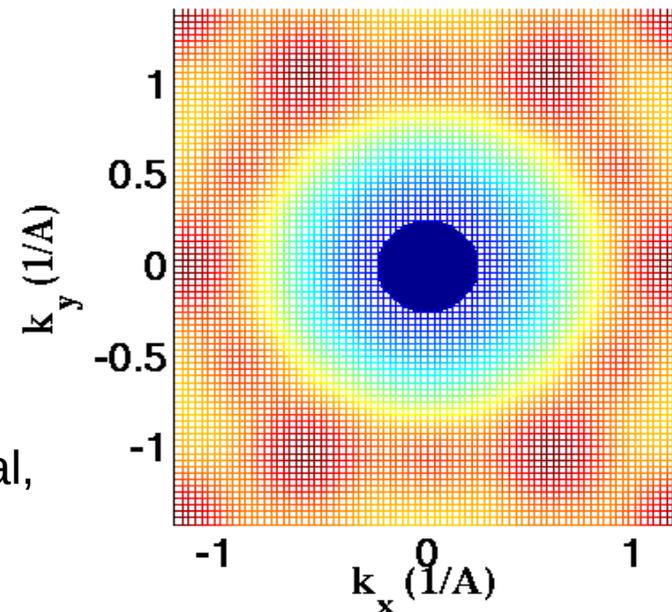
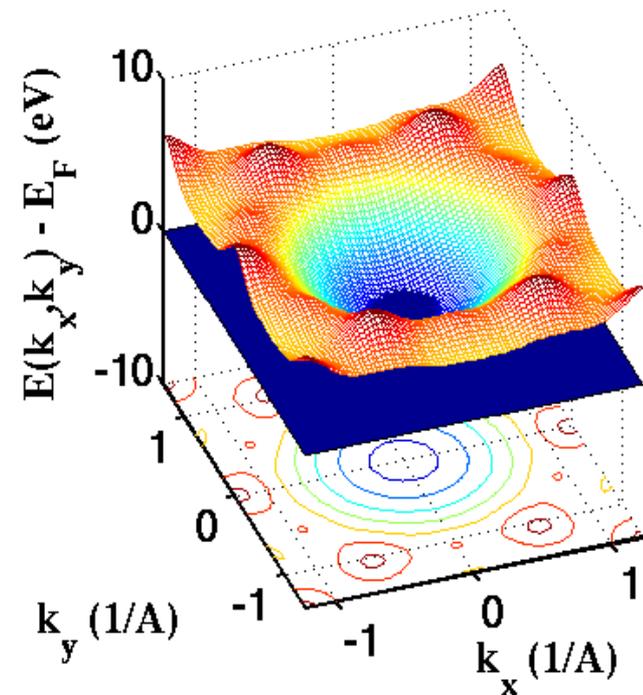


FIG. 2. Energy dispersion relation for the surface state. The solid curve is a parabolic least-squares fit. The shaded region is the projected bulk continuum of states. Note that the surface state enters the bulk continuum just above the Fermi level.



Experimentally observed (ARPES) and calculated (Density Functional Theory + plane-wave + pseudopotential, PWSCF code) surface bands agree well.

Normalized intrinsic emittance

The surface parallel component of the crystal momentum ($\hbar k$) of the electron will be preserved during emission through direct transition.

$E_{kin} = h\nu_{Laser} - \varphi_{work}$: for electrons emitted from the Fermi-level from the center of the Brillouin zone.

$$\beta = \frac{v_e}{c} = \sqrt{1 - \left(\frac{mc^2}{E_{kin} + mc^2} \right)^2}$$

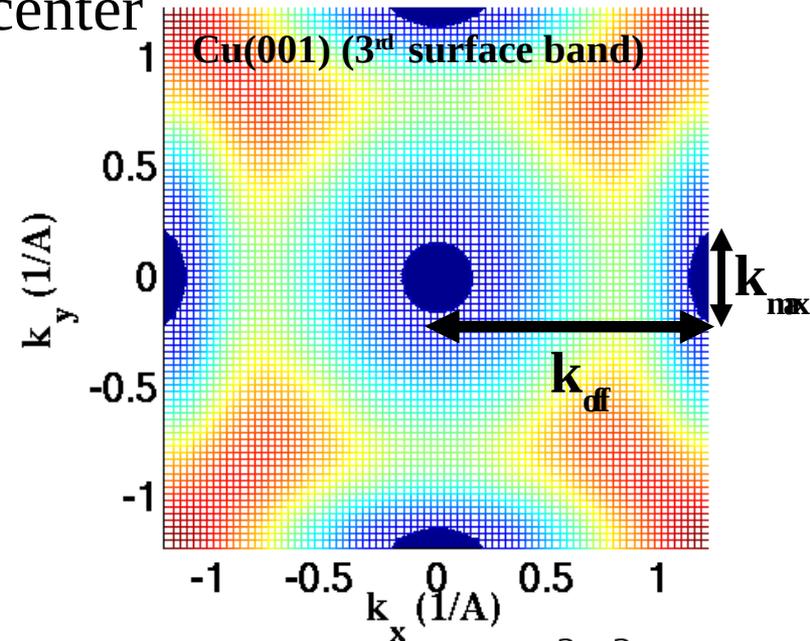
$$p_e = \frac{mv_e}{\sqrt{1 - \beta^2}}$$

$\varepsilon_{nint} = \beta \sigma_x \sigma'_x$: normalized transverse

$\sigma'_x = \arcsin \left(\frac{\sigma_{p_x}}{p_e} \right)$: intrinsic emittance
half the angle of the emission cone

σ_x : radius of Laser beam illumination

$\sigma_{p_x} = \hbar k_{max}$: uncertainty of transverse momentum



$$E_{thermal} = \frac{3 mc^2 \varepsilon_{nint}^2}{2 \sigma_x^2}$$

Table 1: Computed Properties of Various Crystals

Surface *	Computed ϕ (eV)	Experimental ϕ (eV) **	k_{\max} (1/Å)	k_{offset} (1/Å)	E_{th} (eV)	$\epsilon_{n,\text{int}}^{***}$ (mm-mr)
Cu(001)	4.18	4.59	0.15	0	0.27	0.18
Cu(001)	4.18	4.59	0.03	1.229	–	<i>>9.92 eV needed</i>
Cu(111)	4.36	4.94	0.25	0	1.09	0.36
Ag(001)	4.06	4.64	0.10	0	0.12	0.12
Ag(001)	4.06	4.64	0.07	1.087	–	<i>> 8.55 eV needed</i>
Ag(111)	4.17	4.74	0.11	0	0.14	0.13
Thin film	2.92	2.92	0.05	0	0.029	0.06
Thin film	2.92	2.92	0.05	1.087	–	<i>>7.41 eV needed</i>

* Metal (16L) and thin-film (MgO(100)2L-Ag(100)4L-MgO(100)2L) slabs

** Photoemission experimental values for Cu, Ag [20,21]; independent calculation for layered structure [25].

*** Eq. (1); $\sigma_x=0.3$ mm, $h\nu = 4.66$ eV (266 nm) Nd:YAG

Calculations using the PWSCF code with planewaves &DFT PBE (also with PW91) pseudopotentials (norm-preserving and also ultrasoft).



Inspiration from Surface Catalysis: Tuning Work Functions by Oxide Overlayers

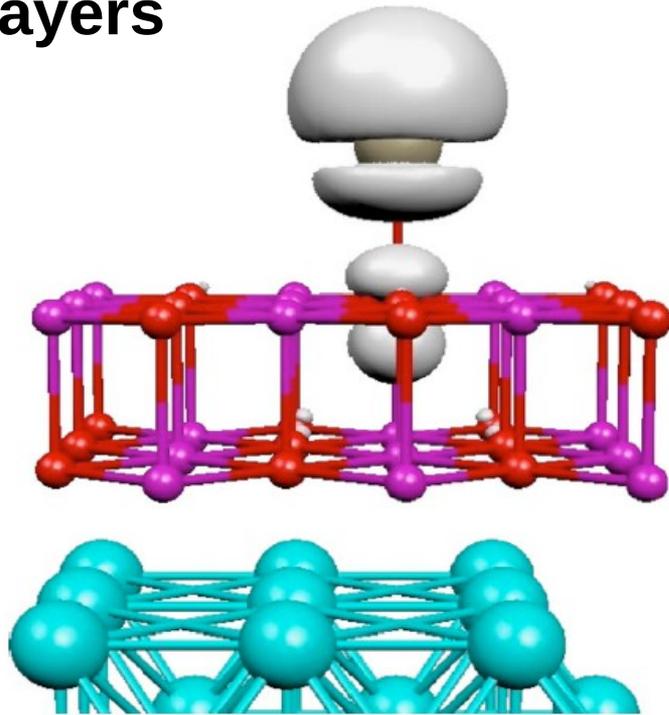


FIG. 5. (Color online) Spin density contour plot for a Au atom adsorbed on the MgO/Ag(100) surface in the absence of substrate relaxation. In this case the charge transfer does not occur and the Au adatom remains essentially neutral.

The catalytic activity (e.g. charging of deposited atoms and clusters) of surfaces is a function of the number of deposited oxide monolayers.

MgO/Ag(001) is one of the most frequently investigated surfaces. Well fitting interface due to close (FCC) lattice parameters: 4.216 Å (MgO) & 4.086 Å (Ag)

Huge work function reduction due to MgO:

$$\Phi_{\text{work}} [\text{Ag}(001)] = 4.64 \text{ eV}$$

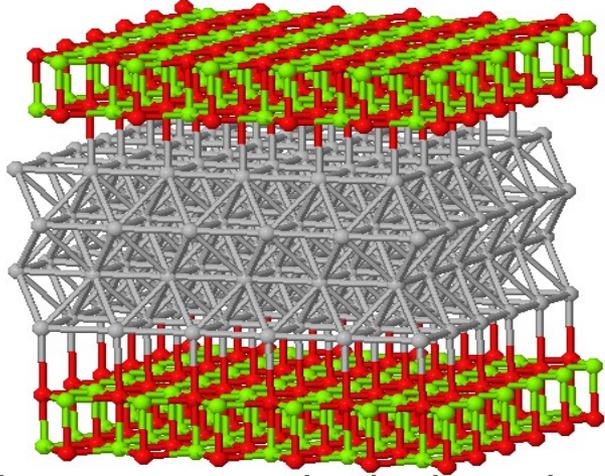
$$\Phi_{\text{work}} [\text{MgO}(2\text{-}8\text{ML})/\text{Ag}(001)] = 2.92 \text{ eV}$$

First predicted, then experimentally measured and confirmed.

Picture from: L. Giordano et.al,
J.Chem.Phys. 127, 144713 (2007)



Can we tune the shape of the surface bands and thus the emittance?



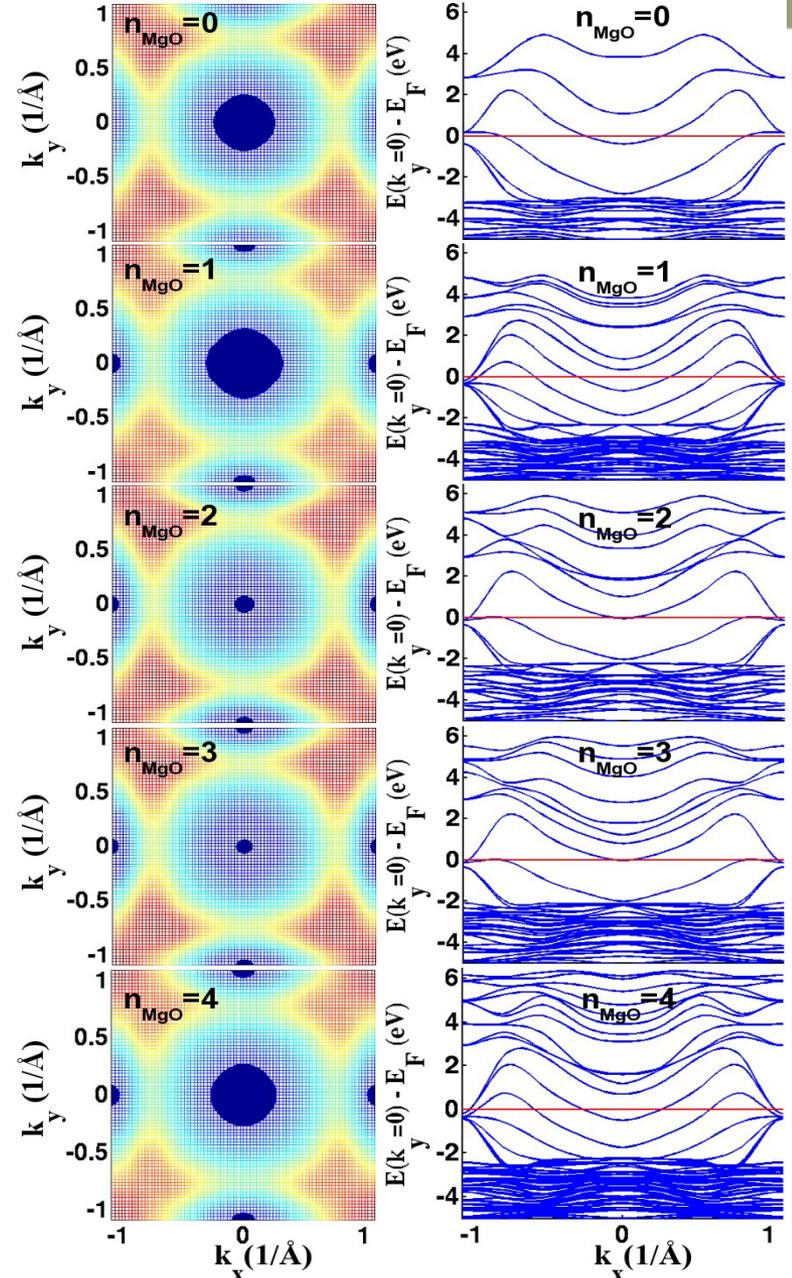
Coloring of atoms:

Mg

O

Ag

The transverse intrinsic emittance is a function of the number of MgO overlayers, with a minimum at $n_{\text{MgO}} = 2, 3$ and with $\epsilon_{\text{ritr}} = 0.06 \text{ mm mrad}$, better than the critical value of $\epsilon_{\text{ritr}} = 0.1 \text{ mm mrad}$ required for new xray sources. Maximum sensitivity to n_{MgO} can be achieved with thin Ag ($\ll 8 \text{ ML}$) and MgO on both top and bottom surfaces. The work function greatly reduces from $\sim 4.6 \text{ eV}$ (Ag) to 2.92 eV (MgO/Ag) due to the MgO overlayers.



K. Németh et.al, Physical Review Letters, 104, 046801 (2010)



Practical Implementation

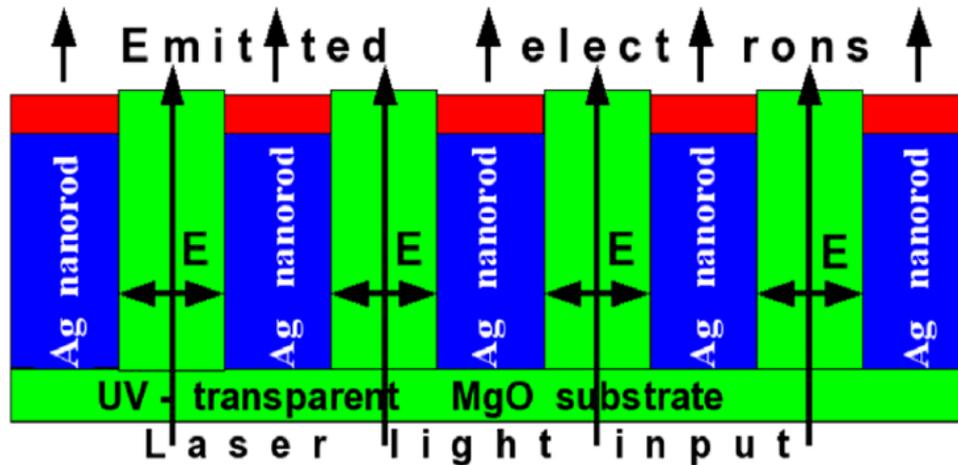


FIG. 3 (color online). Schematic of a photocathode based on a multilayer (red rectangle) topped Ag-nanorod array embedded in a UV-transparent MgO substrate. Typical dimensions of the nanorods are 50–200 nm width and several μm length; the space between the nanorods is commensurate with the width of the nanorods.

Remaining questions:

- Effect of surface roughness and point defects.
- Effect of external E field.
- Stability of surface layers.
- Optimal design of illumination.

Some Basic Nanophotonics Phenomena

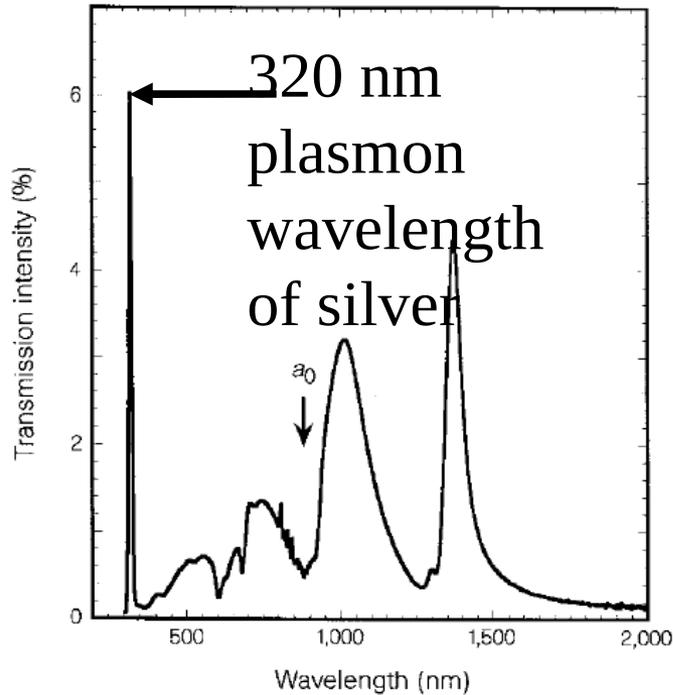


Figure 1 Zero-order transmission spectrum of an Ag array ($a_0 = 0.9 \mu\text{m}$, $d = 150 \text{ nm}$, $t = 200 \text{ nm}$).

t : thickness of metal layer (typically 2-300 nm)
 d : diameter of nanoholes (typically 150 nm)
 a_0 : period of nanoholes (typically $\sim 1 \mu\text{m}$ in a square array)

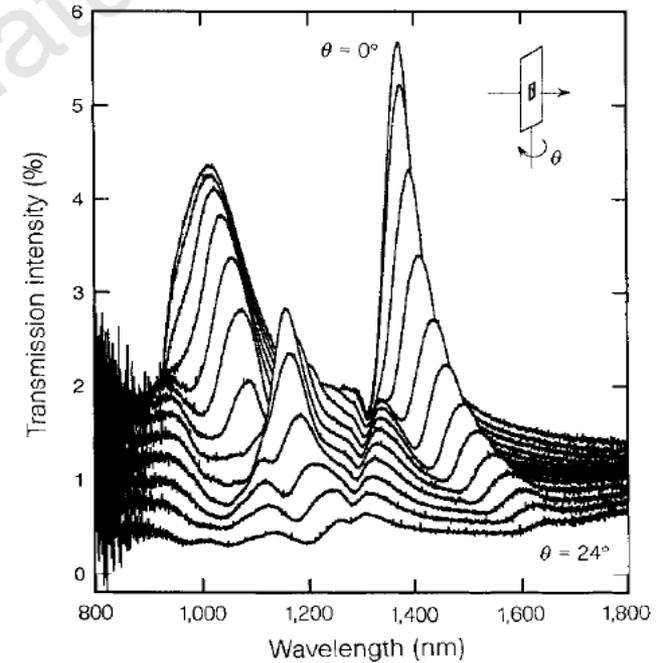


Figure 3 Zero-order transmission spectra as a function of incident angle of the light. Spectra were taken every 2° up to 24° for a square Ag array ($a_0 = 0.9 \mu\text{m}$, $d = 150 \text{ nm}$, $t = 200 \text{ nm}$). The individual spectra are offset vertically by 0.1% from one another for clarity.

Ebbesen, et.al,
Nature, 391, 667 (1998)

Utilization of Nanophotonics Phenomena

Sub-wavelength nanoholes behave similarly to nanorod-arrays and can serve as substrate for backside illumination and higher harmonics Generation. Put MgO over nanoholed Ag layer.

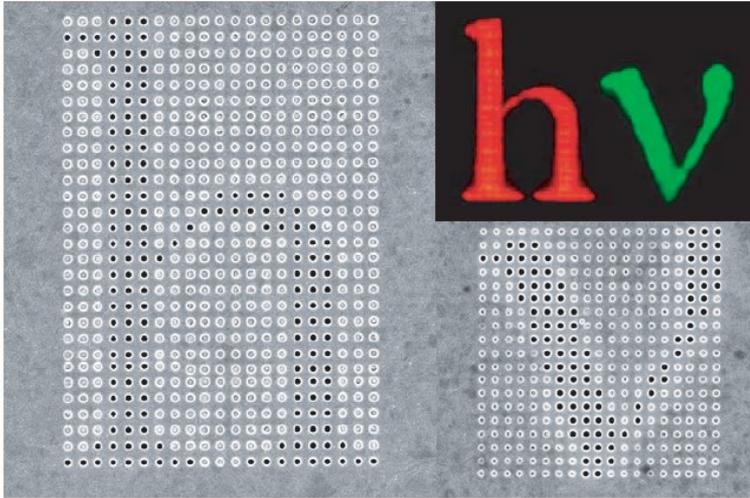


Figure 6 | Holes in a dimple array generating the letters 'hv' in transmission. An array of dimples is prepared by focused-ion-beam milling an Ag film. Some of the dimples are milled through to the other side so that light can be transmitted. When this structure is illuminated with white light, the transmitted colour is determined by the period of the array. In this case the periods were chosen to be 550 and 450 nm respectively to achieve the red and green colours.

C. Genet & T.W. Ebbesen,
Nature 445, p39 (2007)

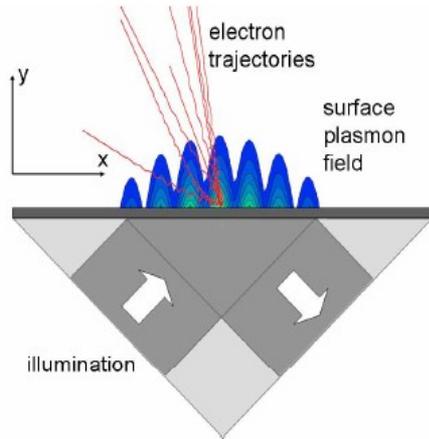
Old illumination scheme:

Start from $\lambda \sim 1\mu\text{m}$ laser wavelength, generate 2nd higher harmonic through photonic crystals to get $\lambda = 266\text{nm}$ light (with great loss of energy), generate required shape of the $\lambda = 266\text{nm}$ pulse (difficult for small wavelength).

Suggested new scheme:

Start from $\lambda \sim 1\mu\text{m}$ laser wavelength, shape pulse in the long wavelength region (easy), illuminate nanoholes from back and use them to generate higher harmonics of $\lambda = 266\text{nm}$ wavelength that will induce electron emission from the top MgO/Ag interface at the other side of the Ag layer.

Utilization of Ponderomotive Acceleration to Generate Bright Electron Beams



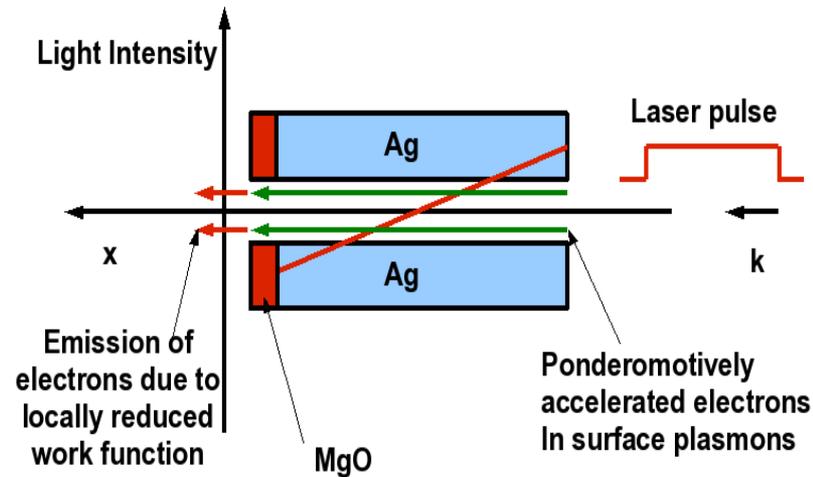
Addition of ultrathin MgO layers would substantially reduce the workfunction, and further improve the brightness of the emitted electron beam.

Fig. 1. The generation of electron beams by SP enhanced electron acceleration. Field amplitudes (false colour plot) and electron trajectories (red lines) illustrate the model used. For further details see text.

Picture from: P. Dombi & P. Racz, Optics Express, 16, 2887 (2008) (simulation of high-brightness beam generation.

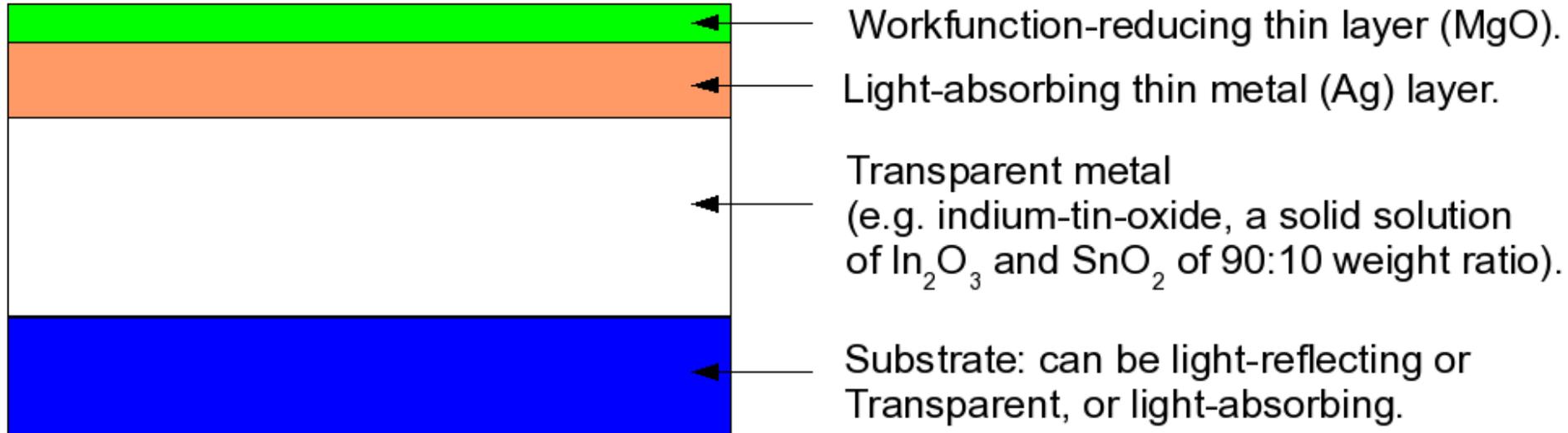
See also:

J. Kuperstych, et.al, "Ponderomotive acceleration of photoelectrons in surface-plasmon-assisted multiphoton photoelectric emission", PRL, 86, 5180 (2001).



Another design based on ponderomotive acceleration of electrons of surface plasmons in a nanohole/nanorod array.

High Brightness Photocathodes Based on Transparent Metals:



The transparent metal provides a conducting support where no electron-excitation will happen, thus electrons won't have to travel to the surface from large distances.

Electron-electron scattering (2nd step of the 3-steps model) will be substantially reduced due to light absorption confined to the thin non-transparent metal coating. The thickness of the light-absorbing metal coating is a small fraction of light's attenuation length in the same metal.

It can be illuminated from the top or from the back-side, or multiple reflection can happen in the transparent metal layer. It can be combined with the Kretschmann arrangement of surface plasmon generation.

CONCLUSION:

**A COMBINATION OF ULTRATHIN OXIDE OVERLAYERS
ON METAL SURFACES
WITH NANOTECHNOLOGY AND NANOPHOTONICS
MAY PAVE THE WAY TOWARD
HIGH BRIGHTNESS ELECTRON AND X-RAY SOURCES.**

THANKS FOR THE ATTENTION!

